

An Improvement of Thermal Conductivity of Underfill Materials for Flip-Chip Packages

Haiying Li, Karl I. Jacob, and C. P. Wong, *Fellow, IEEE*

Abstract—Effective heat dissipation is crucial to enhance the performance and reliability of electronic devices. In this work, the performance of encapsulants filled with carbon fiber was studied and compared with silica filled encapsulants. Encapsulants filled with mixed combination of fillers for optimizing key properties were also investigated. The thermal and electrical conductivities were investigated and glass transition temperature (T_g), thermal expansion coefficient (TCE), and storage modulus (E') of these materials were studied with thermal analysis methods. The composites filled with both carbon fiber and silica showed an increase of thermal conductivity three to five times of that of silica filled encapsulants of the same filler loading while maintaining/enhancing major mechanical and thermal properties.

Index Terms—Carbon fiber, encapsulants, flip-chip packages, glass transition temperature, silica, TCE, underfill materials.

I. INTRODUCTION

THE TREND of developing high speed, high integration and low cost integrated circuits has led to microelectronic components with significant increased internal heat generation. All microelectronics produce heat and a typical microprocessor, especially high performance or high frequency IC, dissipates in excess of 60 W of power today, which is projected to increase to 100 W by 2005. As a result, a higher operating temperature would be expected at steady-state without an effective heat dissipation mechanism [1]. At high operating temperatures the performance and reliability of a component will be decreased exponentially with the temperature. Components are usually protected with polymeric encapsulation in the form of underfilling, potting and glob-top which strongly influence the heat dissipation. The commonly used polymeric encapsulants filled with silica usually exhibit a very poor thermal conductivity (<0.5 W/m.K) as compare to metals. Approaches to increase heat dissipation rate on the component level include providing a heat sink on the back of the component and using fillers of high thermal conductivity in encapsulants. The former has been widely accepted, however, often resulting unavoidably in bulky designs. The later method employs ceramic materials with high thermal conductivity as fillers within the encapsulants. These materials including alumina, boron nitride and aluminum nitride (AlN), as well as their mixtures which show thermal conductivity in the range of 150 to 300 W/m.K [2], [3], and

[4]. Thermal properties of most commonly used materials in electronics and electronics packaging fields are listed in Table I.

Among these materials, diamond is known to have the highest thermal conductivity (2 kW/m.K), however, its high cost largely limits its use as filler in encapsulants. Carbon fibers have a thermal conductivity that can be as high as 800–1200 W/m.K with a negative thermal expansion coefficient (CTE) in their axial direction and provide a strong reinforcement to polymers. However, its electrically conductive property limits its usage in many applications in electronics packaging.

In this study, a carbon fiber of high thermal conductivity was used as the filler in epoxy based underfill materials to enhance the thermal conductivity of underfill materials, while maintaining good electrical insulation of the underfill material.

II. EXPERIMENTAL

A. Materials and Formulation

One grade of carbon fiber provided by BP Amoco Chemicals was used as the thermally conductive filler in this study. This fiber has a diameter of $7\text{ }\mu\text{m}$ and an average length of $45\text{ }\mu\text{m}$. It has an extremely high thermal conductivity, above 800 W/m.K. The scanning electronic microscope (SEM) image and the properties of this fiber are given in Fig. 1 and Table II, respectively. Other fillers used were silica coated aluminum nitride (SCAN) from Dow Chemical Company, and silica from Nippon Chemicals, Inc. The SEM images of these fillers are shown in Figs. 2 and 3, respectively, and their properties are listed in Table II.

Four series of samples were prepared based on a basic formulation technique. A summary of the sample composition is given in Table III. All basic formulations contain two diepoxides, 3, 4-epoxy cyclohexylmethyl-3, 4-epoxy cyclohexyl carboxylate (ERL4221E, EEW 266, Union Carbide), and poly (bisphenol A-co-epichloro-hydrin), glycidyl end-capped (MW377, Aldrich or EPON 8281, Shell), a hardener, hexahydro-4-methylphthalic anhydride (HHMPA) (Lindau Chemicals, Inc.) and a curing catalyst, 1-cyanoethyl-2-ethyl-4-methylimidazole-trimellitate (abbreviated name 2E4MZ-CN, Shikoku Chemicals), and silica powder (LE-03 surface reforming, size $3\text{ }\mu\text{m}$, from Nippon Chemical Industrial Co., Ltd.). All chemicals were used as received without further treatment, and all fillers were dried in a vacuum oven at $125\text{ }^\circ\text{C}$ over night and stored in desiccators.

Table III gives all the formulas used in this study. The stoichiometry of all chemicals used was based on mole, and the addition of each kind of filler was based on the total volume of a formula.

Control samples were prepared by using silica filler in the amount of 0 to 45 volume percent (vol%) with 7.5 vol% interval

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H. Li and K. I. Jacob are with the School of Textile and Fiber Engineering, Georgia Institute of Technology, Atlanta, GA 30332 USA.

C. P. Wong is with the School of Materials Science and Engineering, Electronic Packaging Center, Georgia Institute of Technology, Atlanta, GA 30332 USA (e-mail: cp.wong@mse.gatech.edu).

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TABLE I
THERMAL PROPERTIES OF MATERIALS INVOLVED IN ELECTRONICS PACKAGING [1]

Material	Density (Kg/m ³)	Specific Heat (J/kg·K)	Thermal Conductivity (W/m·K)	Ratio to Air
Air	1.16	1005	0.024	1.0
Epoxy (dielectric)	1500	1000	0.23	9.6
Epoxy (conductive)	10500	1195	0.35	14.6
Polyimide	1413	1100	0.33	13.8
FR-4	1500	1000	0.30	12.5
Water	1000	4200	0.59	24.6
Thermal grease			1.10	46.0
Silica	2200		1.5	62.5
Alumina	3864	834	22	9.17×10^2
Silicon	2330	770	120	5.00×10^3
Aluminum	2700	900	150	6.25×10^3
Aluminum nitride, silica coated	3260		220	9.17×10^3
Boron nitride	2250		300	1.25×10^4
Gold	19300	129	300	1.25×10^4
Copper	8800	380	390	1.63×10^4
Carbon fiber, high thermally conductive	2150-2250		800	3.33×10^4
Diamond	3500	51	2000	8.33×10^4

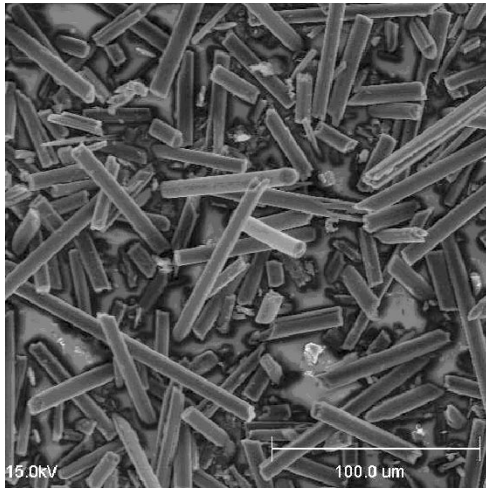


Fig. 1. SEM image of carbon fiber used in this study.

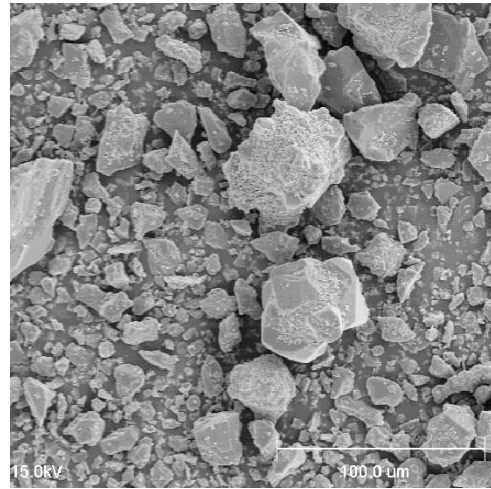


Fig. 2. SEM image of silica coated aluminum nitride used in this study.

between one to another (Sample 1, Table III). In the series of Sample 2, carbon fiber was added as the filler in the amount equal to that of control samples. In the series of Sample 3, both carbon fiber and silica were used as fillers and in the series of Sample 4 both carbon fiber and SCAN powder were added as fillers. In both series the loading level of carbon fiber varied by 7.5 vol% from 0 to 45 vol% while keeping a total filler load of 45 vol%.

All the underfill samples were prepared using the same technique. All chemicals were added and mixed by stirring into a homogeneous mixture in a plastic bottle, and all fillers were mixed into the epoxy resin using a high-speed blender. About

10 g of a formulated material was poured into a glass container and preheated at 100 °C in a vacuum oven over 30 min for de-gassing. Then, the sample was transferred into an aluminum pan of 37.5 mm in diameter, and cured at 130 °C for 30 min, followed by 150 °C for 1 h and 210 °C for another 30 min. After curing the sample was allowed to slowly cool back to room temperature as to avoid the formation of internal stresses. The aluminum pan was peeled off and the sample was polished into a disk with two parallel surfaces (dimension about 35 mm D × 5 mm H) for further analyses. For a specific analysis, a disk sample was usually cut into a required geometry using an Isomet slow speed diamond saw from Buehler.

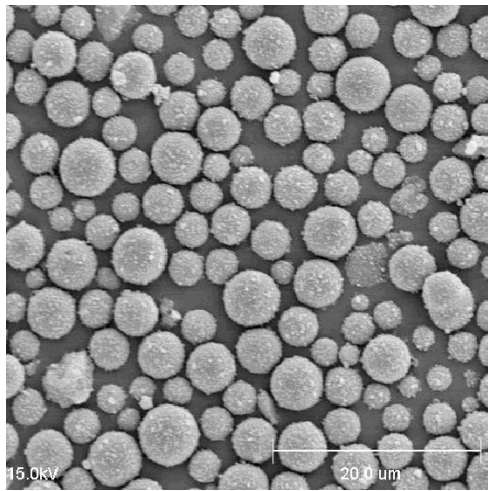


Fig. 3. SEM image of silica used in this study.

B. Measurements of Thermal Conductivity

Solid test samples with a dimension of 25.4×25.4 mm and a thickness varying from 2 to 5 mm were prepared using the technique described in the section of Materials and Formulation. Thermal conductivity was measured at 70°C with a thermal conductance tester TCA-200 from Holometrix Micromet (Metrisc Company). The samples were measured by the guarded heat flow meter method at steady state. A test sample is placed between two plates stabilized at two different temperatures, resulting in a flow of heat from the hotter to the colder plate. The amount of heat is measured with a thin heat flux transducer attached to one of the temperature-controlled plates.

C. Measurements of Electrical Conductivity

A sensitive amperometer, Keithley 485 Autoranging picoammeter, with a dc power supply by Hewlett Packard (6553A DC Power Supply), was used to measure the electrical conductivity at room temperature. Samples were cut into a dimension about $3 \times 5 \times 30$ mm and the electrical conductivity was measured in the longest direction. An electrical conductive adhesive (epoxy filled with silver flakes) tape was used as the end electrodes. The resistivity data were reported in dc resistivity.

D. Thermal Analyses

Thermomechanical Analysis (TMA) was conducted on a TMA (Model 2940 by TA Instruments). The cured sample was heated from 25°C to 250°C in a rate of $5^\circ\text{C}/\text{min}$. and the thickness change versus temperature rise was monitored. The inflection point of thermal expansion was defined as TMA Tg.

Dynamic mechanical analysis (DMA) on dynamic modulus of cured materials was performed on a DMA (Model 2980 by TA Instruments). A specimen for DMA test was a bar of a size about $30 \times 8 \times 3$ mm. The test was performed on a single cantilever under 1 Hz sinusoidal strain loading and the temperature was increased from room temperature to around 250°C in a heating rate of $5^\circ\text{C}/\text{min}$.

III. RESULTS AND DISCUSSION

A. Thermal Conductivity

Carbon fiber with a thermal conductivity as high as $800\text{ W/m}\cdot\text{K}$ was used as filler in epoxy underfill materials. A control formula with silica as the filler was also prepared for the purpose of comparison. As shown in Fig. 4, both silica and carbon fiber filled samples showed a linearly or near linearly increasing thermal conductivity as filler loading increases. The thermal conductivity of the carbon fiber filled sample increased from 0.42 to $1.82\text{ W/m}\cdot\text{K}$ as the filler loading level varies from 15 to 45 vol%, 2 to 4.5 times of those of silica filled control samples at the same filler loading level.

Carbon fiber was also used together with silica as co-fillers in a series of underfill materials with a constant total filler load of 45 vol%. The choosing of this volume ratio was based on the observation that further filling of fillers beyond this point caused difficulty in material mixing and more voids in the samples. As seen in Fig. 5, when the carbon fiber loading varied from 7.5 to 37.5 vol%, the thermal conductivity increased from 1.3 to $2.8\text{ W/m}\cdot\text{K}$, three to seven times of that of 45 vol% silica filled sample. Also noted is that the thermal conductivity of all samples in this series were higher than those of corresponding carbon fiber filled samples. This is understandable when the carbon fiber loading was less than 45 vol% because the addition of silica, higher in thermal conductivity than the epoxy resin, for 45 vol% total filler composition replaces epoxy which has a lower thermal conductivity. In the case of 45 vol% of filler loading however, that the thermal conductivity of the co-filler filled sample was higher than carbon fiber only filled one (Figs. 4 and 5) is intriguing. An explanation could be the mixing of carbon fiber and silica formed a structure of higher packing density and thus provided a better thermal conductive pathway than carbon fiber filler only.

Aluminum nitride has a thermal conductivity over $300\text{ W/m}\cdot\text{K}$. Because of its high thermal conductivity and low toxicity, it has been subjected to extensive study since late 1980's to determine its suitability in electronic packaging applications. In the present study SCAN powder was used together with carbon fiber as the co-filler to make a total filler load of 45 vol%. At 0 vol% of carbon fiber loading or, in the other word, 45 vol% loading of SCAN, the thermal conductivity was $1.48\text{ W/m}\cdot\text{K}$. As 7.5 vol% of carbon fiber was added the thermal conductivity was enhanced significantly to $1.87\text{ W/m}\cdot\text{K}$ (Fig. 5). However, further increasing of carbon fiber filling level showed a slightly decrease in thermal conductivity, and when the carbon fiber level increased to 37.5 vol%, the thermal conductivity was only $1.2\text{ W/m}\cdot\text{K}$. It was probably due to the severe sedimentation of SCAN caused by its high density ($>3.5\text{ g/cm}^3$) resulting in the formation of a polymer resin rich top layer after curing, which is more thermal insulating than the fillers.

TABLE II
PROPERTIES OF EPOXY RESIN, CARBON FIBER AND OTHER FILLERS USED

Properties	Epoxy	Silica	SCAN	Carbon Fiber
Average Diameter (micron)		3	75	7
Ave. Filament Length (micron)				45
Density (g/cm ³)	1.16	2.2	3.26	2.15-2.25
Surface Area (m ² /g)				0.4
Thermal Conductivity (W/m·K)	0.20	1.5	220	>800 (axial)
Electrical Resistivity (Ohm·m)				<3.0×10 ⁻⁶
CTE at Room Temperature (ppm/°C)	88	0.5	4.4	-1.45 (axial)
Young's Modulus (GPa)	2.25	73	330	
Bulk Modulus (GPa)	3.75	39	220	
Tensile Modulus (GPa)				(6.89-9.30)×10 ²
Tensile Strength (GPa)				>1.38
Poisson's Ratio	0.19	0.19	0.25	

TABLE III
FORMULATIONS USED IN THIS STUDY

Formula Component	Sample Series 1	Sample Series 2	Sample Series 3	Sample Series 4
ERL-4221E (mol.)	1	1	1	1
Bisphenol-A (mol.)	1	1	1	1
HHMPA (mol.)	1.6	1.6	1.6	1.6
EMZCN (Catalyst, mol.)	0.03	0.03	0.03	0.03
Silica, (vol.%)	0/7.5/15/22.5/30/37.5/45		45/37.5/30/22.5/15/7.5	
Carbon fiber (vol.%)		0/7.5/15/22.5/30/37.5/45	0/7.5/15/22.5/30/37.5	0/7.5/15/22.5/30/37.5
AlN, glass coated (vol.%)				45/37.5/30/22.5/15/7.5
Total filler content (vol.%)	0/7.5/15/22.5/30/37.5/45	0/7.5/15/22.5/30/37.5/45	45	45

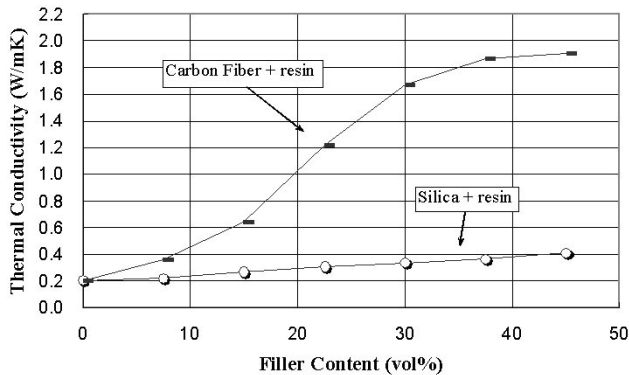


Fig. 4. Thermal conductivity of carbon fiber and silica filled systems.

B. Electrical Conductivity

From the application point of view, underfill materials must be electrical insulating. Since the carbon fiber used is somewhat electrical conductive (Table II), the electrical conductivity of the

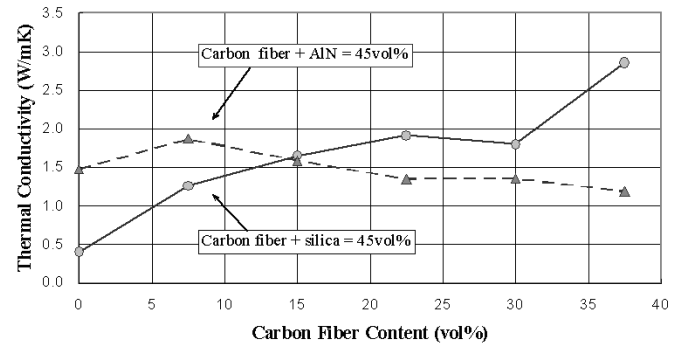


Fig. 5. Thermal conductivity of carbon fiber and silica, and carbon fiber and silica coated aluminum nitride filled systems. The total filler content were constantly 45vol%.

materials was a key concern. The electrical dc resistivity of all samples was measured as described in the experimental section and the results are shown in Fig. 6. The electrical resistivity of both carbon fiber filled, and carbon fiber/silica-filled samples kept a high and stable resistivity about 10^8 Ohm·m, the

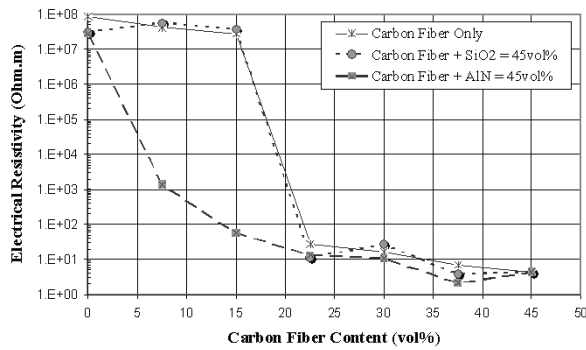


Fig. 6. Electrical resistivity of sample series 2, 3, and 4.

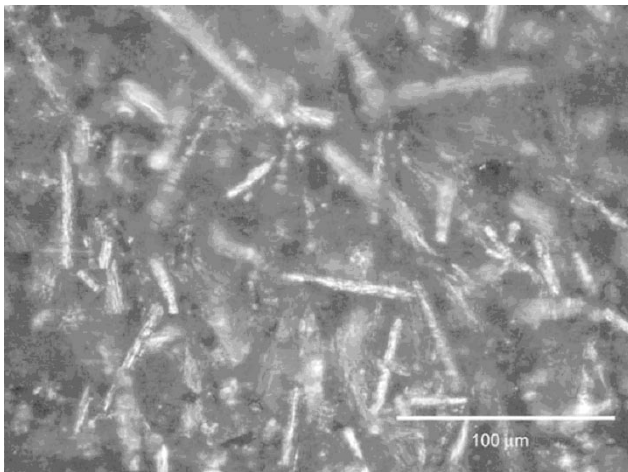


Fig. 7. Microscopic image of the sample containing 7.5 vol.% of carbon fiber and 37.5 vol.% of silica.

same order of magnitude as that of both silica and SCAN filled samples when the carbon fiber loading was below 15 vol%. As the loading of carbon fiber was higher than 15 vol%, the resistivity quickly dropped to $10 \Omega\text{m}$ level, and it did not change too much as the carbon fiber load increased further. In other words, a threshold existed between 15 to 22.5 vol% filler loading. On the other hand, the resistivity of carbon fiber/SCAN filled samples exhibited a dramatic decrease to the order of $10^{-1} \Omega\text{m}$, as the carbon fiber loading increasing. The difference in the resistivity change pattern of these two series of samples probably was due to the separation of SCAN and carbon fiber caused by the large difference in density and particle size of these two fillers. During epoxy resin curing process, the viscosity of the samples decrease dramatically and denser filler will sediment to the bottom faster than the lighter one. In a multi-filler filled system, this may lead to filler separation. When particle sizes of fillers are quite different, the large filler may not distribute well into the smaller filler. In any cases the smaller filler, carbon fiber, may form a continuous phase which is more electrical conductive (Figs. 7 and 8).

The results of this test indicate that in order to obtain a good insulation of an underfill material, the loading level of carbon fiber must be less than 15 vol% in carbon fiber/silica filled underfills, and 7.5 vol% or even less in carbon fiber/SCAN filled

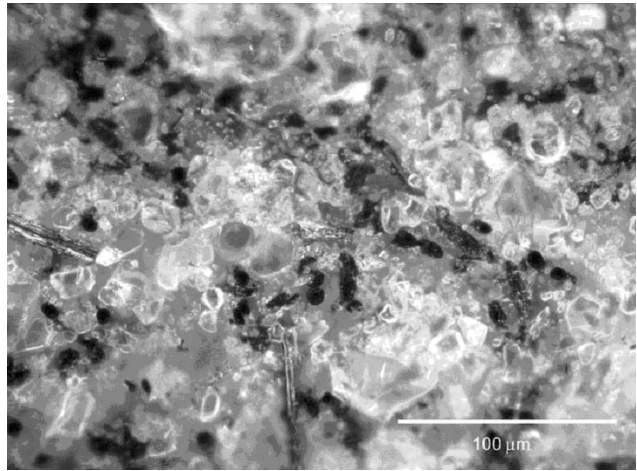


Fig. 8. Microscopic image of the sample containing 7.5 vol.% of carbon fiber and 37.5 vol.% of SCAN.

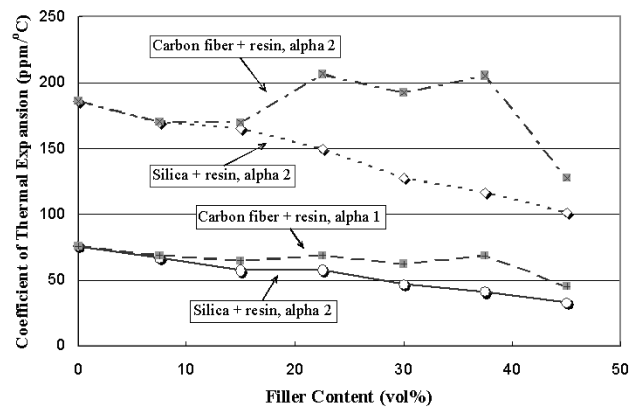


Fig. 9. CTE of sample series 1 and 2.

underfills. From Fig. 5, one can find that the thermal conductivity of Sample Series 3 is about $1.65 \text{ W/m}\cdot\text{K}$, which is about three times of that of the 45 vol% silica filled control sample.

C. Thermal Analyses

Thermo-mechanical properties including glass transition temperature, coefficient of thermal expansion, as well as storage and loss moduli are critical properties for underfill materials. In order to observe these parameters, TMA and DMA were conducted on all samples. The CTE data are shown in Figs. 9 and 10 and DMA results are given in Figs. 11–13. In CTE diagrams all CTE observed below T_g were assigned as $\alpha 1$ and those above T_g were labeled as $\alpha 2$.

Figs. 9 and 11 show the CTE's and modulus of silica filled and carbon fiber filled samples, respectively. As the filler content was increased, $\alpha 1$ of both types of samples and $\alpha 2$ of silica series decreased but the modulus increased for both types of samples. However, $\alpha 2$ of carbon fiber filled samples fluctuated up and down about the $\alpha 2$ of unfilled sample. As comparison, $\alpha 1$ and $\alpha 2$ of each carbon fiber samples were higher than its counterpart of the same filler loading. $\alpha 1$ of carbon fiber samples was averagely 1.56 times of that of silica samples with the

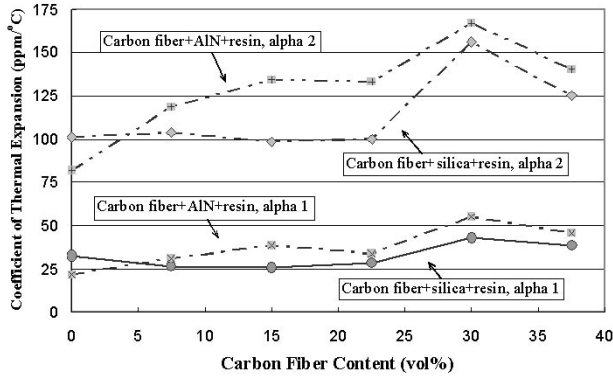


Fig. 10. CTE of sample series 3 and 4.

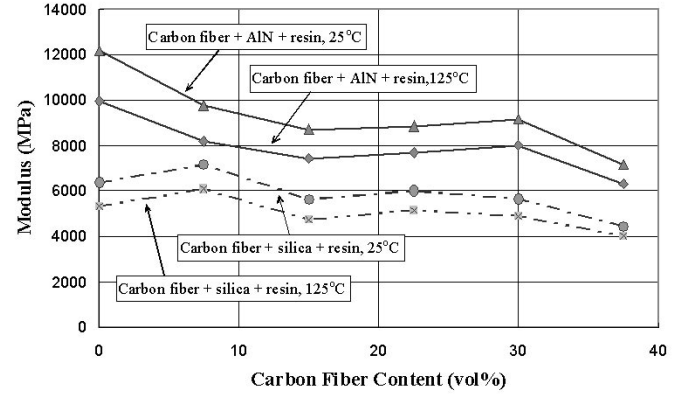


Fig. 12. Storage modulus of sample series 3 and 4.

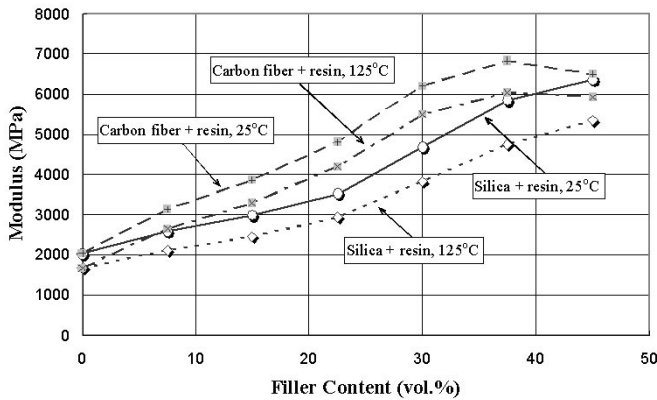


Fig. 11. Storage modulus of sample series 1 and 2.

lowest ratio (1.12) located at 15 vol% loading level. Modulus diagrams interpreted from DMA of these two series (Fig. 11) show that almost all the storage modulus of carbon fiber filled materials, both at 25 and 125 °C, were about 800 MPa higher than that of their silica filled counterparts. This indicates a stronger reinforcement of carbon fiber to the composite.

In the case of the two series of underfill materials using dual fillers (Figs. 10, 12, and 13), the 45vol% of silica and carbon fiber, and 45 vol% of SCAN and carbon fiber filled samples showed slightly increased α_1 from 32.6 to 38.5 ppm/m°C and from 21.9 to 45.7 ppm/m°C, respectively as the carbon fiber ratio increased with a constant total filler load of 45 vol%. The α_2 of silica based samples generally showed lower values than that of corresponding SCAN based samples. As the carbon fiber content increased, the α_2 of the series of Sample 3 increased from 101 to 156 and then dropped to 125 ppm/m°C, correspondingly, the α_2 of SCAN based samples varied from 82 to 167 and dropped to 141 ppm/m°C.

When the carbon fiber was not added, the 45 vol% of silica filled sample showed storage moduli of 6.5 GPa at 25 °C and 5.3 GPa at 125 °C whereas the 45 vol% of SCAN filled sample exhibited extremely high moduli: 12 GPa at 25 °C and 10 GPa at 125 °C. Generally, as the carbon fiber content increased, the

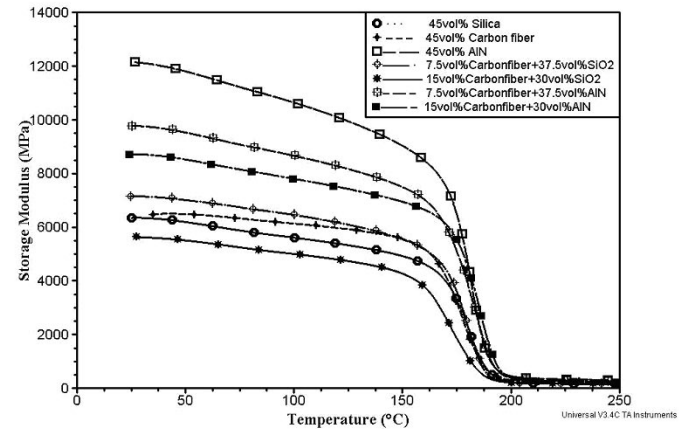


Fig. 13. DMA diagram of seven selected samples.

storage moduli of both series at both 25 °C and 125 °C slightly decreased and the storage moduli at 25 °C were about 1 GPa higher than that at 125 °C for both series of samples. As the carbon fiber content increased to 7.5 vol%, the moduli of the silica based series increased about 0.7–0.8 GPa whereas those of the SCAN based system decreased about 2 GPa at both 25 °C and 125 °C. The further increase in carbon fiber ratio did not change the moduli of both systems very much at both temperatures, except the samples containing 37.5 vol% of carbon fiber in both series, in which case voids were seen on the sample surfaces caused by the high viscosity before curing. Fig. 13 shows the DMA diagrams of seven selected samples that exhibited the high moduli at room temperature.

Glass transition temperatures of these four series of samples observed on TMA and DMA were shown in Figs. 14 and 15. In general, the Tgs of both silica and carbon fiber filled systems varied between 150 to 180 °C, and no obvious trend can be followed. The carbon fiber filled system showed slightly higher values than the former when filler loading was low. At high filler loading levels however, this trend reversed. The Tgs of carbon fiber-silica-resin system fluctuated between 160 to 180 °C with the highest point at 7.5 vol% carbon fiber content. For carbon

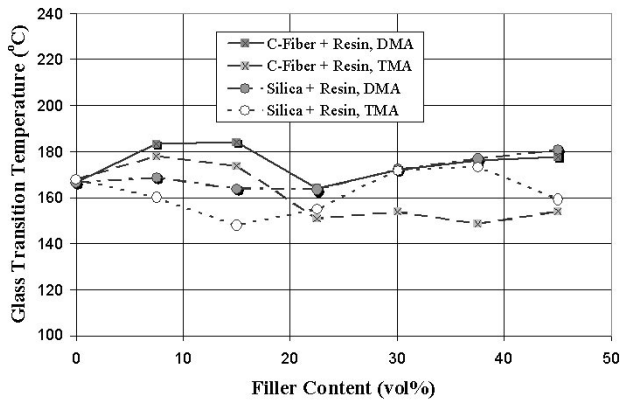


Fig. 14. Glass transition temperatures of carbon fiber and silica filled systems.

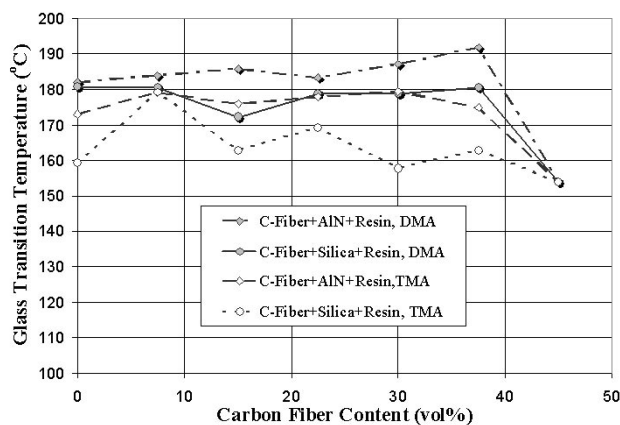


Fig. 15. Glass transition temperatures of carbon fiber/silica and carbon fiber/SCAN filled systems.

fiber-SCAN-resin system, Tgs observed on TMA and DMA varied at 170 to 180 °C, and 180 to 190 °C, respectively.

Based on the results of thermo-mechanical and thermal conductance studies one may conclude that lightly carbon fiber filled SCAN based and silica based samples are promising candidates of high thermally conductive underfill materials. However, the low electrical resistivity of carbon fiber containing SCAN based samples excludes them from the application as underfill materials. The samples containing 7.5 vol% and 15 vol% of carbon fiber in the carbon fiber/silica filled system on the other hand showed fairly high electrical resistivity and storage moduli comparable to those of silica filled samples, as well as high thermal conductivity of 1.27 and 1.66 W/m·K, respectively. These thermal conductivity data are about 300% of that of 45 vol% silica filled material, even though not as high as that of the 7.5 vol% carbon fiber/SCAN filled sample. These samples are excellent candidates of high thermally conductive underfill materials.

IV. CONCLUSION

Carbon fiber with high thermally conductivity was used as a filler to enhance the thermal conductivity of epoxy resin-based

underfill materials and the major properties related to the applications were investigated. Four series of underfill samples containing silica, carbon fiber, carbon fiber combined with silica, and carbon fiber combined with aluminum nitride were studied for their thermal conductivity, electrical conductivity and thermo-mechanical properties. The underfill filled with 37.5 vol% carbon fiber showed a thermal conductivity of 1.9 W/m·K, a value five times that of silica-filled underfill. Carbon fiber provided a more enhanced storage modulus and comparable CTE to underfill materials as compared to silica filler. When both carbon fiber and silica fillers were used, a thermal conductivity as high as 2.86 W/m·K was observed on the sample with 37.5 vol% of carbon fiber. Also in this system, the samples containing 7.5 vol% and 15% of carbon fiber showed thermal conductivity of 1.27 and 1.66 W/m·K, respectively. These represent a 300% enhancement from that of 45 vol% silica filled material with comparable storage moduli. Electrical resistance study showed that when carbon fiber content was no more than 15 vol%, a fairly high electrical resistivity, comparable to that of silica filled underfill, was maintained on all systems except the system containing both carbon fiber and SCAN. In summary, samples containing 7.5 vol% of carbon fiber and 37.5 vol% of silica, and 15 vol% of carbon fiber and 30 vol% silica are excellent candidates of high thermally conductive underfill materials for high performance flip-chip packaging applications.

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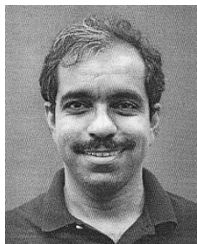
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Haiying Li received the B.S. degree in polymer science and engineering from the Beijing Institute of Chemical Technology, Beijing, China, in 1983, the M.S. degree in chemistry from Clark College, Atlanta, GA, in 1996, and is currently pursuing the Ph.D. degree at the Georgia Institute of Technology, Atlanta.

After receiving the B.S. degree, he served as a Polymer Engineer in industry for ten years. He has authored and co-authored 16 journal and proceedings articles and three U.S. patents.

Mr. Li received the Third Award of the Light Industry Ministry of China for the Progress in Science and Technology, in 1989 and 1991 and was nominated as one of the finalists for ICI Student Award in Applied Polymer Science in 2000. He is a member of the American Chemical Society.



Karl I. Jacob received the B.Sc. degree in civil engineering from Kerala University, Trivandrum, India, in 1978, the M.Tech. degree in applied mechanics from the Indian Institute of Technology, Madras, in 1980, and the Ph.D. degree in engineering mechanics from the Ohio State University, Columbus, in 1985.

He is currently an Associate Professor of the School of Textile and Fiber Engineering, Georgia Institute of Technology, Atlanta. He teaches graduate and undergraduate courses and conducts research in polymer physics and engineering, rheology, and mechanics of polymeric materials. His graduate work was in the area of numerical analysis of vibrating three-dimensional structures. Previously, he was with the Dacron Research Laboratory, DuPont Company, and later at the Experimental Station (Pioneering Research Laboratory/Central Research and Development).

Dr. Jacob is the Chair of the Polymer Technical Committee, American Society of Mechanical Engineers (ASME), as well as the Chair of the Textile Engineering Division, ASME.



C. P. Wong (SM'87–F'92) received the B.S. degree in chemistry from Purdue University, West Lafayette, IN, and the Ph.D. degree in organic/inorganic chemistry from Pennsylvania State University, University Park.

After his doctoral study, he was awarded two years as a Postdoctoral Scholar at Stanford University, Stanford, CA. He joined AT&T Bell Laboratories, in 1977 as Member of Technical Staff. He was elected an AT&T Bell Laboratories Fellow in 1992. He is a Regents Professor with the School of Materials

Science and Engineering and a Research Director at the NSF-funded Packaging Research Center, Georgia Institute of Technology, Atlanta. He holds over 45 U.S. patents, numerous international patents, has published over 400 technical papers and 300 key-notes and presentations in the related area. His research interests lie in the fields of polymeric materials, high T_c ceramics, materials reaction mechanism, IC encapsulation, in particular, hermetic equivalent plastic packaging, electronic manufacturing packaging processes, interfacial adhesions, PWB, SMT assembly, and components reliability.

Dr. Wong received the AT&T Bell Laboratories Distinguished Technical Staff Award in 1987, the AT&T Bell Labs Fellow Award in 1992, the IEEE Components, Packaging and Manufacturing Technology (CPMT) Society Outstanding and Best Paper Awards in 1990, 1991, 1994, 1996, and 1998, the IEEE Technical Activities Board Distinguished Award in 1994, the 1995 IEEE CPMT Society's Outstanding Sustained Technical Contribution Award, the 1999 Georgia Tech's Outstanding Faculty Research Program Development Award, the 1999 NSF-Packaging Research Center Faculty of the Year Award, the Georgia Tech Sigma Xi Faculty Best Research Paper Award, the University Press (London, UK) Award of Excellence, and was elected a member of the National Academy of Engineering in 2000. He is a Fellow of AIC and AT&T Bell Labs. He served as the Technical Vice President (1990 and 1991), and the President (1992 and 1993) of the IEEE-CPMT Society.